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14. ABSTRACT

Cavity-enhanced direct frequency comb spectroscopy combines broad bandwidth, high spectral resolution, and ultrahigh detection sensitivity in one experimental platform based on an optical frequency comb efficiently coupled to a high-finesse cavity. The effective interaction length between light and matter is increased by the cavity, massively enhancing the sensitivity for measurement of optical losses. Individual comb components act as independent detection channels across a broad spectral window, providing rapid parallel processing. In the past year we have further developed and refined this JILA-pioneered technology, and implemented the first applications that demonstrate the enormous potential of this spectroscopic method. In particular, we have developed various frequency comb sources, techniques for efficient coupling between comb and cavity, and detection schemes that utilize the technique's high-resolution, wide-bandwidth, and fast data-acquisition capabilities. Optical frequency comb-based high-resolution spectrometers offer enormous potential for spectroscopic applications.

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Final report to AFOSR on FA9550-09-1-0085 “Production, Detection, and Control of Ultracold Molecules via Optical Frequency Combs”
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Cavity-enhanced direct frequency comb spectroscopy combines broad bandwidth, high spectral resolution, and ultrahigh detection sensitivity in one experimental platform based on an optical frequency comb efficiently coupled to a high-finesse cavity. The effective interaction length between light and matter is increased by the cavity, massively enhancing the sensitivity for measurement of optical losses. Individual comb components act as independent detection channels across a broad spectral window, providing rapid parallel processing. In the past year we have further developed and refined this JILA-pioneered technology, and implemented the first applications that demonstrate the enormous potential of this spectroscopic method. In particular, we have developed various frequency comb sources, techniques for efficient coupling between comb and cavity, and detection schemes that utilize the technique’s high-resolution, wide-bandwidth, and fast data-acquisition capabilities.

Optical frequency comb−based high−resolution spectrometers offer enormous potential for spectroscopic applications. However, the lack of suitable mid - infrared comb sources has impeded explorations of molecular fingerprinting. We have now demonstrated for the first time a frequency−comb Fourier transform spectrometer operating in the 2100−to−3700 cm^{-1} spectral region that allows fast and simultaneous acquisitions of broadband absorption spectra with up to 0.0056 cm^{-1} resolution. We demonstrate part−per−billion detection limits in 30 seconds of integration time for various important molecules including methane, ethane, isoprene, and nitrous oxide. Our system enables precise concentration measurements even in gas mixtures that exhibit continuous absorption bands, and it allows detection of molecules at levels below the noise floor via simultaneous analysis of multiple spectral features. This system represents a near real−time, high−resolution, high−bandwidth mid−infrared spectrometer which is ready to replace traditional Fourier transform spectrometers for many applications in trace gas detection, atmospheric science, and medical diagnostics.

Cavity-enhanced direct frequency comb spectroscopy (CE-DFCS) has demonstrated powerful potential for trace gas detection based on its unique combination of high bandwidth, rapid data acquisition, high sensitivity, and high resolution, which is unavailable with conventional systems. However, previous demonstrations have been limited to proof-of-principle experiments or studies of fundamental laboratory science. We have now demonstrated the development of CE-DFCS towards an industrial application – measuring impurities in arsine, an important process gas used in III-V semiconductor compound manufacturing. A strongly absorbing background gas with an extremely complex, congested, and broadband spectrum renders trace detection exceptionally difficult, but the capabilities of CE-DFCS overcome this challenge and make it possible to identify and quantify multiple spectral lines associated with water impurities. Further, frequency combs allow easy access to new spectral regions via efficient nonlinear optical processes. In the work supported by AFOSR we demonstrate

detection of multiple potential impurities across 1.75-1.95 μm (5710-5130 cm^{-1}) with single-channel detection sensitivities of 1×10^{-7} cm^{-1} $Hz^{-1/2}$ in nitrogen and identify water doped in arsine with a sensitivity of 1×10^{-6} cm^{-1} $Hz^{-1/2}$.

With the AFOSR support, we have also recently achieved the objective of quantum-noise-limited optical frequency comb spectroscopy. Direct frequency comb spectroscopy, first reported in Science in 2004, combines properties that were previously considered exclusive, i.e., wide spectral bandwidth, high optical resolution, and fast acquisition time. However, so far the sensitivity of the technique has been limited by technical noise. For example, when a dispersive element and multidetector array are used to analyze the comb spectrum transmitted through the gas sample, the sensitivity is limited by detector noise because of the low optical power in each resolved spectral element. Higher sensitivities can be achieved by an implementation of a high-finesse external cavity, in which the interaction length of the light with the sample, and thus the absorption signal, is drastically increased. The equidistant spectrum of the comb matches that of an external cavity thus enabling efficient coupling of laser power into the cavity. However, the narrow cavity modes convert laser frequency noise to amplitude noise. Several detection schemes have been developed to overcome this issue, but again none was able to achieve the fundamental detection limit set by the shot noise. In our recent work we achieved for the first time shot-noise-limited sensitivity with cavity-enhanced direct frequency comb spectroscopy. The obtained minimum detectable absorption of $1.7 \times 10\text{-}12$ cm^{-1} per spectral element at 400 s of acquisition time is so far the lowest for a frequency-comb-based spectroscopic technique. We achieved this record-high sensitivity by combining a frequency comb tightly locked to a high-finesse cavity and a fast-scanning Fourier transform spectrometer with auto-balancing dual beam detection. We emphasize that we demonstrate shot-noise-limited sensitivity not only in background spectra but also in properly normalized absorption spectra. We also prove that the technique is capable of long-term stable operation, while and a single spectrum is acquired in times as short as 1 second, depending on the resolution. The system operates at low as well as atmospheric pressures. The high signal-to-noise ratio and the high stability of the system enable analysis of absorption lineshapes, which are in excellent agreement with theoretical model. This unprecedented level of control in comb spectroscopy has allowedenables multi-line fitting over the entire whole available spectral bandwidth available from the comb, unleashing the full potential of cavity-enhanced direct frequency comb spectroscopy. The shot-noise-limited system achieved in our system represents the ultimate implementation of cavity-enhanced direct frequency comb spectroscopy. We expect the technique to have a significant impact in many areas of molecular spectroscopy for applications such as medical breath analysis, detection of pollutants and hazardous gases, astrocombs or spectroscopy of ultracold molecules. Furthermore, this work can trigger applications in comb-based quantum information processing, which requires the signal to be limited only by the noise of the photons.

Finally, we have recently demonstrated the first cavity-enhanced optical frequency comb spectroscopy in the mid-infrared wavelength range. The system, aimed at highly sensitive detection of hydrogen peroxide, is based on a mid-infrared optical parametric oscillator synchronously pumped by a high power Yb:fiber laser, a high-finesse broadband cavity,

and a fast-scanning Fourier transform spectrometer with auto-balancing detection. A comb spectrum with a bandwidth of 150 nm centered around 3.7 μm is simultaneously coupled to the cavity and both degrees of freedom of the OPO comb are stabilized to the cavity to ensure stable transmission. The auto-balancing detection scheme reduces the intensity noise by a factor of *250*, and a sensitivity of 6.9×10^{-11} cm^{-1} $Hz^{-1/2}$ per spectral element with a resolution of 800 MHz is achieved (6000 elements, 5.4×10^{-9} cm^{-1} $Hz^{-1/2}$ for a single element). This corresponds to a detection limit for hydrogen peroxide of 8 parts-per-billion (ppb). Spectra of acetylene, methane and nitrous oxide at atmospheric pressure are also presented, and a lineshape model is developed and used to fit to the experimental data.